

## WHAT IS CLAIMED IS:

1. A method of independently producing an independent negative electrode by itself, wherein said negative electrode includes a thin film of an inorganic solid electrolyte and is suitable for use in a lithium secondary cell, wherein said method comprises the following steps in sequence:
  - a) providing a plurality of containers including at least one closed container containing a source material for said inorganic solid electrolyte and containing a negative electrode base material having a surface made of at least one of lithium metal and lithium alloys;
  - b) placing said at least one closed container into at least one inlet chamber space insulated from air;
  - c) opening said at least one closed container in said at least one inlet chamber space, and taking out said base material and said source material from said at least one container in said at least one inlet chamber space;
  - d) transferring said base material and said source material, without exposure to air, from said at least one inlet chamber space into a film forming apparatus that is adjacent and connected in an airtight manner to said at least one inlet chamber space;
  - e) in said apparatus, carrying out a film forming process using said source material to form said thin film of said inorganic solid electrolyte on said base material, to thereby make said independent negative electrode including said base material with said thin film formed thereon;
  - f) transferring said independent negative electrode, without exposure to air, from said film forming apparatus into an outlet chamber space that is insulated from air and is adjacent and connected in an airtight manner to said film forming apparatus;
  - g) in said outlet chamber space, without exposing said independent negative electrode to air, placing said independent negative electrode into a storage

28 container selected from among said plurality of containers, and closing  
29 said storage container; and

30 h) removing said storage container, with said independent negative electrode  
31 closed therein, from said outlet chamber space into an environment of  
32 atmospheric air.

1 2. The method according to claim 1, wherein said at least one inlet chamber space  
2 and said outlet chamber space is respectively substantially inactive to lithium.

1 3. The method according to claim 1, wherein said outlet chamber space is separate  
2 and distinct from said at least one inlet chamber space.

1 4. The method according to claim 1, wherein said outlet chamber space is the same  
2 chamber space as one of said at least one inlet chamber space.

1 5. The method according to claim 1, wherein said storage container is separate and  
2 distinct from said at least one closed container.

1 6. The method according to claim 1, wherein said storage container is the same  
2 container as one of said at least one closed container being reused as said storage  
3 container.

1 7. The method according to claim 1, further comprising, after said step h), taking  
2 said independent negative electrode out of said storage container, and assembling  
3 said independent negative electrode with other components to make a lithium  
4 secondary cell.

1 8. The method according to claim 1, before said step a) further comprising a  
2 preliminary step of making said negative electrode base material by forming a

thin film of said at least one of lithium metal and lithium alloys to form said surface on a substrate material by a vapor deposition process.

**9.** The method according to claim 8, comprising carrying out said vapor deposition process so as to form said thin film with a thickness of at most 20  $\mu\text{m}$  on said substrate material.

**10.** The method according to claim 1, further comprising, during said steps c) and d), filling said at least one inlet chamber space and said film forming apparatus with a gas selected from the group consisting of helium, nitrogen, neon, argon, krypton, a mixture gas of at least two of the foregoing gases, and dry air having a dew point of  $-50^{\circ}\text{C}$  or below.

**11.** The method according to claim 1, further comprising, during said steps f) and g), filling said outlet chamber space and said film forming apparatus with a gas selected from the group consisting of helium, nitrogen, neon, argon, krypton, a mixture gas of at least two of the foregoing gases, and dry air having a dew point of  $-50^{\circ}\text{C}$  or below.

**12.** The method according to claim 1, wherein said thin film of said inorganic solid electrolyte has a composition containing: 30 to 65 atomic percent of lithium; sulfur; and at least one element selected from the group consisting of phosphorous, silicon, boron, germanium, and gallium.

**13.** The method according to claim 12, wherein said composition further contains at least one of oxygen and nitrogen.

**14.** The method according to claim 12, wherein said thin film of said inorganic solid electrolyte is amorphous.

1     **15.**    The method according to claim 12, wherein said thin film of said inorganic solid  
2            electrolyte has an ionic conductance of at least  $1 \times 10^{-4}$  S/cm at 25°C.

1     **16.**    The method according to claim 12, wherein said film forming process is a  
2            process selected from the group consisting of sputtering, vapor evaporation,  
3            laser ablation, and ion plating.

1     **17.**    A method of independently producing an independent negative electrode by  
2            itself, wherein said negative electrode includes a thin film of an inorganic solid  
3            electrolyte and is suitable for use in a lithium secondary cell, wherein said  
4            method comprises the following steps in sequence:

5            a)    providing a plurality of containers including at least one closed container  
6                   containing a first source material of at least one of lithium metal and  
7                   lithium alloys, and containing a second source material for use in forming  
8                   said inorganic solid electrolyte;

9            b)    placing said at least one closed container into at least one inlet chamber  
10                  space insulated from air;

11           c)    opening said at least one closed container in said at least one inlet  
12                  chamber space, and taking out said first source material and said second  
13                  source material from said at least one container in said at least one inlet  
14                  chamber space;

15           d)    transferring said first source material and said second source material,  
16                  without exposure to air, from said at least one inlet chamber space into  
17                  a film forming apparatus that is adjacent and connected in an airtight  
18                  manner to said at least one inlet chamber space;

19           e)    in said apparatus, carrying out a first film forming process using said first  
20                  source material to form a base thin film of said first source material on  
21                  a base material, and carrying out a second film forming process using

22 said second source material to form said thin film of said inorganic solid  
23 electrolyte on said base thin film on said base material, to thereby make  
24 said independent negative electrode including said base material with said  
25 thin film formed thereon;

26 f) transferring said independent negative electrode, without exposure to air,  
27 from said film forming apparatus into an outlet chamber space that is  
28 insulated from air and is adjacent and connected in an airtight manner to  
29 said film forming apparatus;

30 g) in said outlet chamber space, without exposing said independent negative  
31 electrode to air, placing said independent negative electrode into a storage  
32 container selected from among said plurality of containers, and closing  
33 said storage container; and

34 h) removing said storage container, with said independent negative electrode  
35 closed therein, from said outlet chamber space into an environment of  
36 atmospheric air.

1 **18.** The method according to claim 17, wherein said at least one inlet chamber space  
2 and said outlet chamber space is respectively substantially inactive to lithium.

1 **19.** The method according to claim 17, wherein said outlet chamber space is separate  
2 and distinct from said at least one inlet chamber space.

1 **20.** The method according to claim 17, wherein said outlet chamber space is the  
2 same chamber space as one of said at least one inlet chamber space.

1 **21.** The method according to claim 17, wherein said storage container is separate  
2 and distinct from said at least one closed container.

1     **22.**    The method according to claim 17, wherein said storage container is the same  
2            container as one of said at least one closed container being reused as said storage  
3            container.

1     **23.**    The method according to claim 17, further comprising, after said step h), taking  
2            said independent negative electrode out of said storage container, and assembling  
3            said independent negative electrode with other components to make a lithium  
4            secondary cell.

1     **24.**    The method according to claim 17, wherein said first film forming process is a  
2            vapor deposition process.

1     **25.**    The method according to claim 24, comprising carrying out said vapor  
2            deposition process so as to form said base thin film with a thickness of at most  
3            20  $\mu\text{m}$  on said base material.

1     **26.**    The method according to claim 17, further comprising, during said steps c) and  
2            d), filling said at least one inlet chamber space and said film forming apparatus  
3            with a gas selected from the group consisting of helium, nitrogen, neon, argon,  
4            krypton, a mixture gas of at least two of the foregoing gases, and dry air having  
5            a dew point of  $-50^{\circ}\text{C}$  or below.

1     **27.**    The method according to claim 17, further comprising, during said steps f) and  
2            g), filling said outlet chamber space and said film forming apparatus with a gas  
3            selected from the group consisting of helium, nitrogen, neon, argon, krypton,  
4            a mixture gas of at least two of the foregoing gases, and dry air having a dew  
5            point of  $-50^{\circ}\text{C}$  or below.

1     **28.**     The method according to claim 17, wherein said thin film of said inorganic solid  
2             electrolyte has a composition containing: 30 to 65 atomic percent of lithium;  
3             sulfur; and at least one element selected from the group consisting of  
4             phosphorous, silicon, boron, germanium, and gallium.

1     **29.**     The method according to claim 28, wherein said composition further contains  
2             at least one of oxygen and nitrogen.

1     **30.**     The method according to claim 28, wherein said thin film of said inorganic solid  
2             electrolyte is amorphous.

1     **31.**     The method according to claim 28, wherein said thin film of said inorganic solid  
2             electrolyte has an ionic conductance of at least  $1 \times 10^{-4}$  S/cm at 25°C.

1     **32.**     The method according to claim 28, wherein said second film forming process is  
2             a process selected from the group consisting of sputtering, vapor evaporation,  
3             laser ablation, and ion plating.

1     **33.**     A method of independently producing an independent negative electrode by  
2             itself, wherein said negative electrode includes a thin film of an inorganic solid /  
3             electrolyte and is suitable for use in a lithium secondary cell, wherein said  
4             method comprises the following steps:

- 5             a)     providing a first closed container containing a first source material  
6                     selected from the group consisting of lithium metal and lithium alloys;
- 7             b)     placing said first closed container into a first inlet chamber space  
8                     insulated from air;
- 9             c)     opening said first closed container in said first inlet chamber space, and  
10                    taking out said first source material from said first closed container in  
11                    said first inlet chamber space;

- 12 d) transferring said first source material, without exposure to air, from said  
13 first inlet chamber space into a first film forming apparatus that is  
14 adjacent and connected in an airtight manner to said first inlet chamber  
15 space;
- 16 e) in said first film forming apparatus, carrying out a first film forming  
17 process using said first source material to form a first thin film of said  
18 first source material on a base material provided in said first film forming  
19 apparatus, to make an intermediate component including said first thin  
20 film on said base material;
- 21 f) transferring said intermediate component, without exposure to air, from  
22 said first film forming apparatus into a first outlet chamber space that is  
23 insulated from air and is adjacent and connected in an airtight manner to  
24 said first film forming apparatus;
- 25 g) in said first outlet chamber space, without exposure to air, placing said  
26 intermediate component into a temporary storage container, and closing  
27 said temporary storage container;
- 28 h) providing a second closed container containing a second source material  
29 for use in forming said inorganic solid electrolyte;
- 30 i) placing said temporary storage container and said second closed container  
31 into a second inlet chamber space insulated from air;
- 32 j) opening said temporary storage container and said second closed  
33 container in said second inlet chamber space, and taking out said  
34 intermediate component and said second source material from said  
35 temporary storage container and said second container in said second inlet  
36 chamber space;
- 37 k) transferring said intermediate component and said second source material,  
38 without exposure to air, from said second inlet chamber space into a  
39 second film forming apparatus that is adjacent and connected in an  
40 airtight manner to said second inlet chamber space;



- 41 l) in said second film forming apparatus, carrying out a second film forming  
42 process using said second source material to form said thin film of said  
43 inorganic solid electrolyte on said intermediate component, to thereby  
44 make said independent negative electrode including said base material  
45 with said first thin film and said thin film of said inorganic solid  
46 electrolyte formed thereon;
- 47 m) transferring said independent negative electrode, without exposure to air,  
48 from said second film forming apparatus into a second outlet chamber  
49 space that is insulated from air and is adjacent and connected in an  
50 airtight manner to said second film forming apparatus;
- 51 n) in said second outlet chamber space, without exposing said independent  
52 negative electrode to air, placing said independent negative electrode into  
53 a storage container and closing said storage container; and
- 54 o) removing said storage container, with said independent negative electrode  
55 closed therein, from said second outlet chamber space into an  
56 environment of atmospheric air.

1 **34.** The method according to claim 33, wherein all of said chamber spaces are  
2 substantially inactive to lithium.

1 **35.** The method according to claim 33, wherein said first outlet chamber space is  
2 separate and distinct from said first inlet chamber space, and said second outlet  
3 chamber space is separate and distinct from said second inlet chamber space.

1 **36.** The method according to claim 33, wherein said first outlet chamber space is the  
2 same chamber space as said first inlet chamber space, and said second outlet  
3 chamber space is the same chamber space as said second inlet chamber space.

1     **37.**     The method according to claim 33, wherein said storage container is separate  
2             and distinct from said temporary storage container.

1     **38.**     The method according to claim 33, wherein said storage container is the same  
2             container as said temporary storage container being reused as said storage  
3             container.

1     **39.**     The method according to claim 33, further comprising, after said step o), taking  
2             said independent negative electrode out of said storage container, and assembling  
3             said independent negative electrode with other components to make a lithium  
4             secondary cell.

1     **40.**     The method according to claim 33, wherein said first film forming process is a  
2             vapor deposition process.

1     **41.**     The method according to claim 40, comprising carrying out said vapor  
2             deposition process so as to form said first thin film with a thickness of at most  
3             20  $\mu\text{m}$  on said base material.

1     **42.**     The method according to claim 33, further comprising, during said steps c) and  
2             d) and during said steps j) and k), respectively filling said first and second inlet  
3             chamber spaces and said first and second film forming apparatuses with a gas  
4             selected from the group consisting of helium, nitrogen, neon, argon, krypton,  
5             a mixture gas of at least two of the foregoing gases, and dry air having a dew  
6             point of  $-50^{\circ}\text{C}$  or below.

1     **43.**     The method according to claim 33, further comprising, during said steps f) and  
2             g) and during said steps m) and n), respectively filling said first and second  
3             outlet chamber spaces and said first and second film forming apparatuses with

a gas selected from the group consisting of helium, nitrogen, neon, argon, krypton, a mixture gas of at least two of the foregoing gases, and dry air having a dew point of -50°C or below.

**44.** The method according to claim 33, wherein said thin film of said inorganic solid electrolyte has a composition containing: 30 to 65 atomic percent of lithium; sulfur; and at least one element selected from the group consisting of phosphorous, silicon, boron, germanium, and gallium.

**45.** The method according to claim 44, wherein said composition further contains at least one of oxygen and nitrogen.

**46.** The method according to claim 44, wherein said thin film of said inorganic solid electrolyte is amorphous.

**47.** The method according to claim 44, wherein said thin film of said inorganic solid electrolyte has an ionic conductance of at least  $1 \times 10^{-4}$  S/cm at 25°C.

**48.** The method according to claim 44, wherein said second film forming process is a process selected from the group consisting of sputtering, vapor evaporation, laser ablation, and ion plating.